Fission Yields and the Oklo Reactor

Radioactivity - Radionuclides - Radiation
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Case Study: Fission Yield Module and the Oklo Nuclear Geyser

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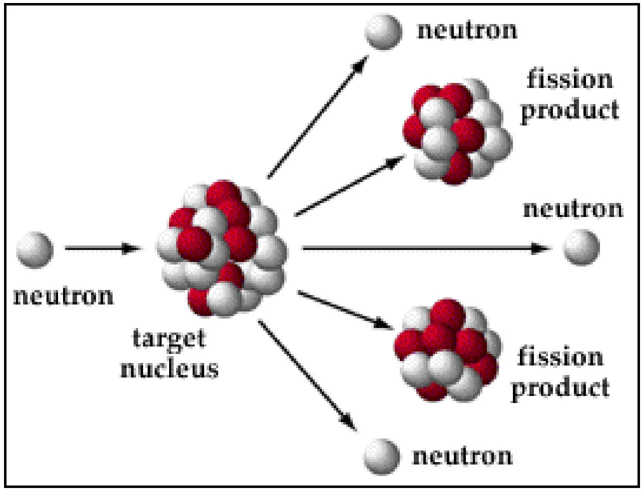
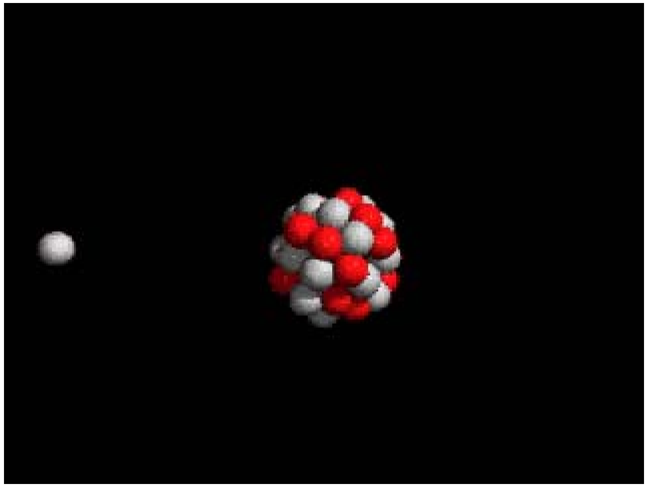
Neutron Induced Fission

- Neutron transport calculations are complicated; one must resort to Monte Carlo techniques which are computationally expensive.
- A tool for analysis of neutron induced fission is included in Nuclides.net: the fission yield module.
- It can be used to calculate the relative abundances of the various fission products for fissile radioisotopes.





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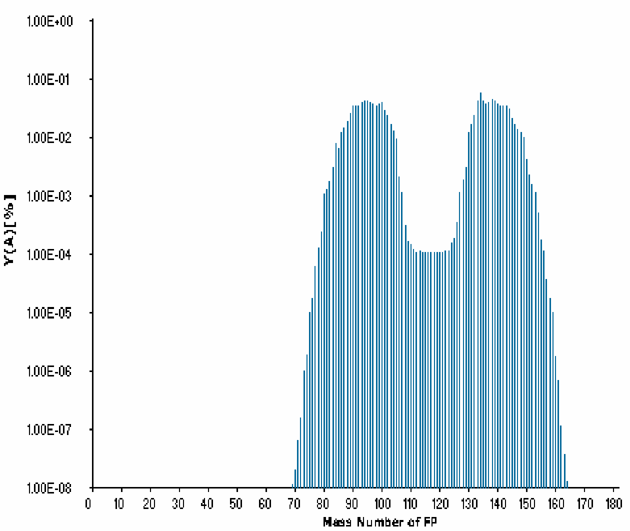
Fission Yields

- Independent yield: number of atoms of a specified nuclide produced directly (after emission of prompt neutrons but excluding radioactive decay) per fission
- Cumulative yield: number of atoms of a specific nuclide produced directly and via decay of precursors per fission
- Chain yield: number of isobars of specific mass produced per fission

Fission Products

- Isotopes of more than 30 elements are observed as fission products
- Most of the fragments are far from stability and decay by β^- or delayed neutron emission



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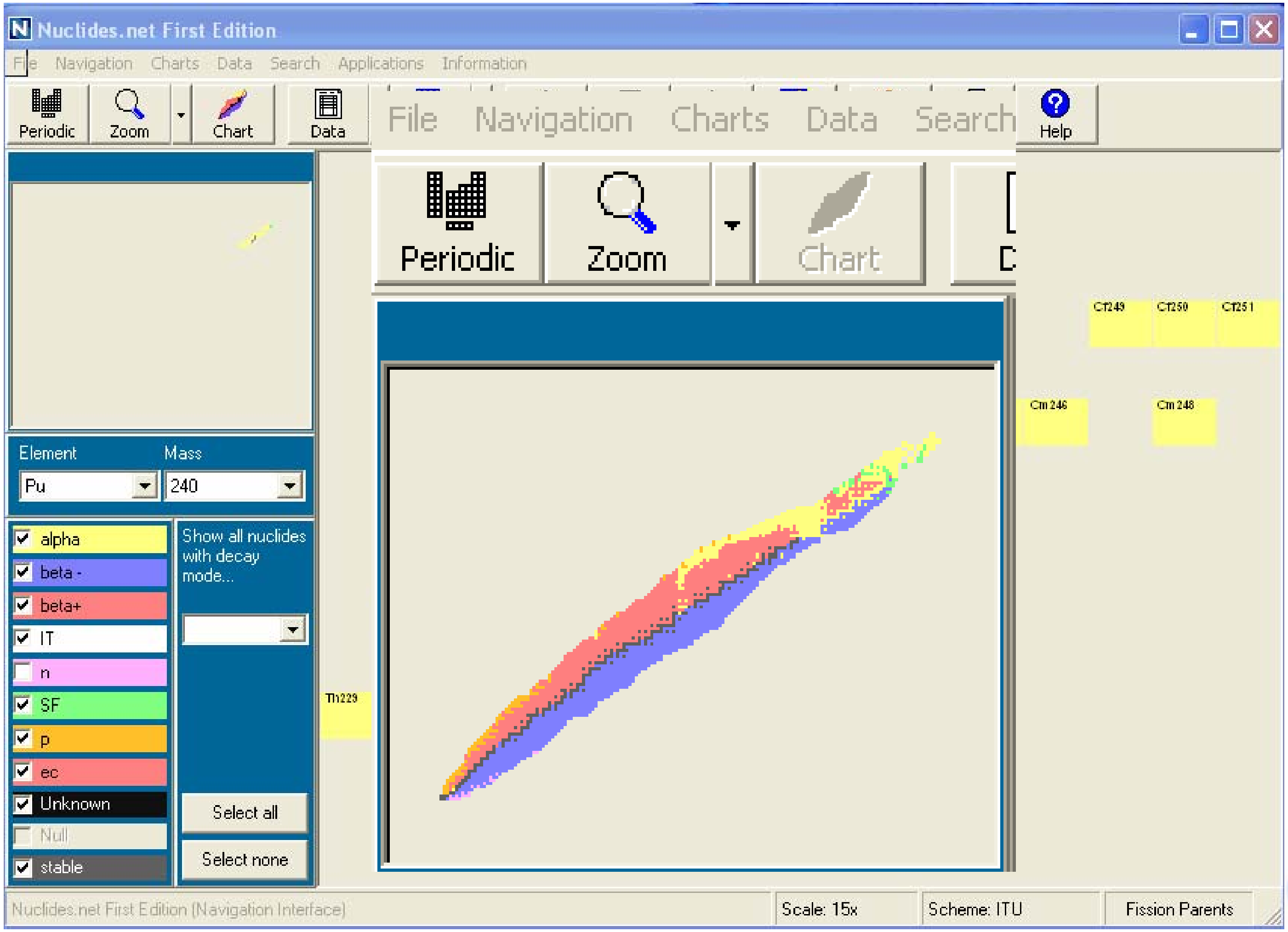
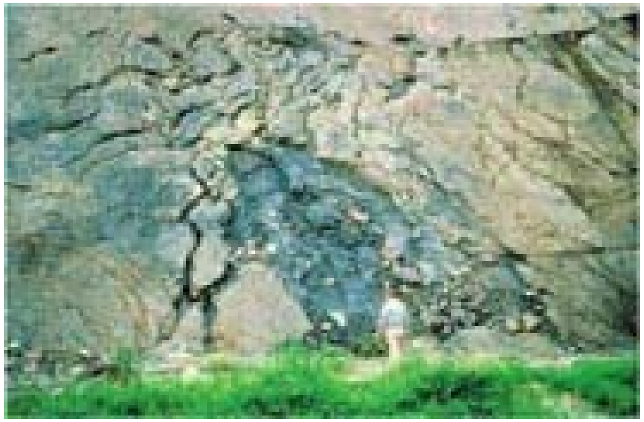
Fission Yield Data in Nuclides.net



Discovery of the Oklo Reactor

- In 1972, uranium ore from French mines at the Oklo site in Gabon (in central Africa) was found to be depleted in ^{235}U.
- Something of a mystery: isotopic composition is globally homogeneous (^{235}U abundance = 0.72%).
- Some process other than radioactive decay must be responsible for this depletion.
- Crucially, fission products and higher actinides were found amongst the depleted ore.
- ***Amazingly, this was the first evidence of naturally occurring nuclear fission chain reactions, a phenomenon first proposed in 1956 by Kuroda.***



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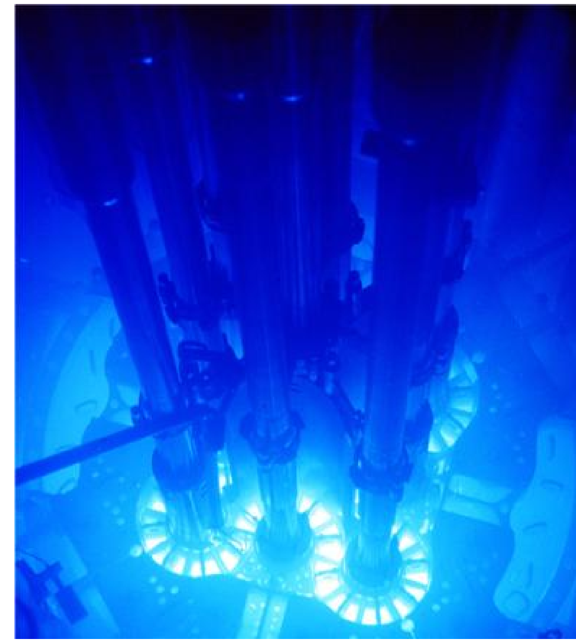
Kuroda's Conditions for Natural Chain Reaction

1. Size of uranium deposit must be larger than 0.7m.
2. Neutron moderator must be present.
3. ^{235}U must be present in sufficient abundance.

- Natural uranium fuel (0.72% ^{235}U) in today's power reactors require heavy water or graphite moderators to thermalise neutrons and sustain chain reaction.
- 1.8 billion years ago, ^{235}U enrichment was around 3%
- This means that a light water (i.e. groundwater) moderator is sufficient for sustained nuclear fission (c.f. present-day LWR).



ATR, US DoE, Idaho National Lab

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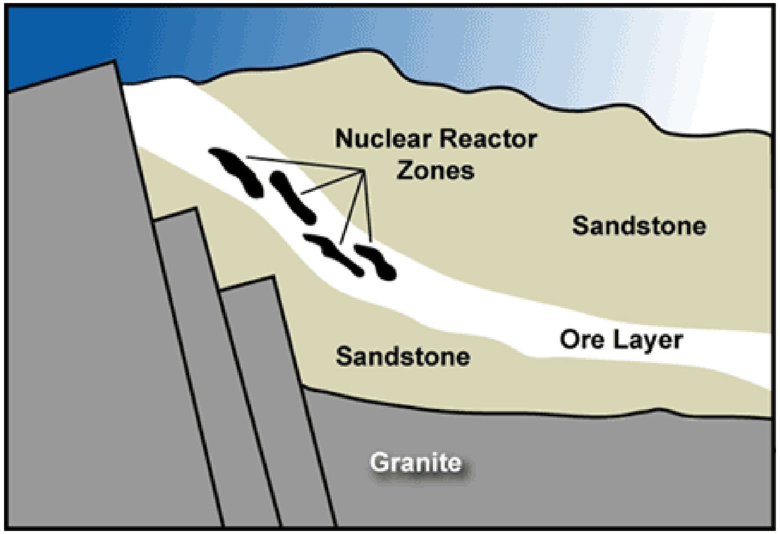
US DoE, Office of Civilian Radioactive Waste Management



APS, Physics Central

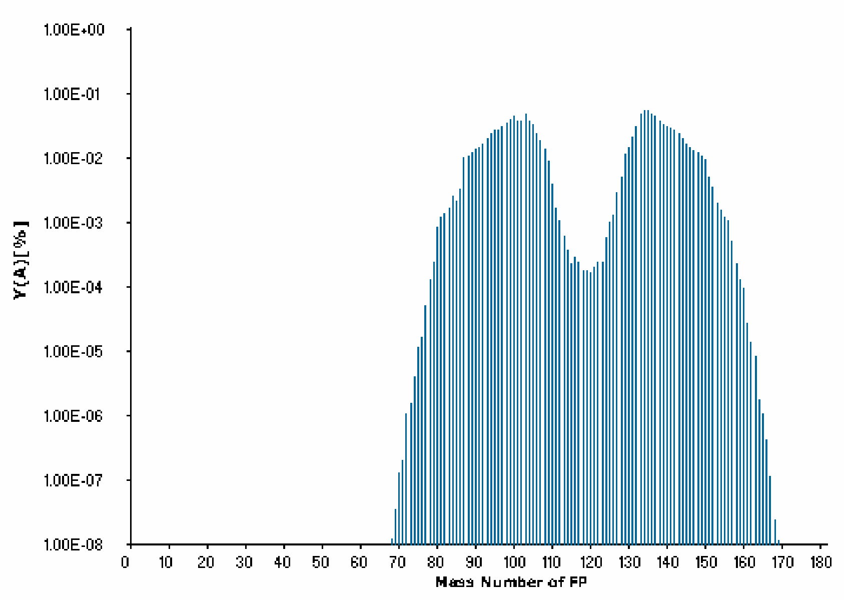
Natural Reactor Properties

- 16 individual reactor zones found, each with slightly different properties.
- Operated around 1.8 billion years ago.
- 15 GWyr of energy released over a period of 150 000 years.
- Corresponds to an average power of 100 kW.
- Over 5 t of uranium consumed.
- Typical neutron fluence around 10^{21} ncm^{-2}

- How do we know about these properties today?
Analysis of Fission Products …

Analysis of Fission Products

- Fission products serve as an ***isotopic fingerprint*** for the types of nuclear processes which occurred at Oklo almost 2 billion years ago.
- Today, we measure the abundance of stable/very long-lived isotopes in samples taken from the Oklo mines.
- Analysis of the Oklo samples involves the three primary nuclear processes: fission, transmutation (e.g. n capture) and radioactive decay.
- Can deduce reactor properties such as age, lifetime, fuel, neutron fluence, average power output and specific characteristics of operational cycle.



Analysis of Xenon Composition (I)

- Xenon is an extremely rare noble gas: well-suited to studies of nuclear reactions.
- Oklo sample analysed via laser mass spectroscopy by Meshik and collaborators at Washington University, St Louis.
- Three amazing results:
  - Largest Xe concentrations ever found in any natural material.
  - Xe not located in Uranium ore; rather in neighbouring Aluminium Phosphates.
  - Isotopic composition of Xe in $AlPO_4$ is different from typical fission yield.
- Detailed calculations have shown that this Xe must have arisen from decay of short-lived fission products such as I, Te and Sb which were captured in the $AlPO_4$ crystals.
- ***What does this tell us about the natural reactor operation?***

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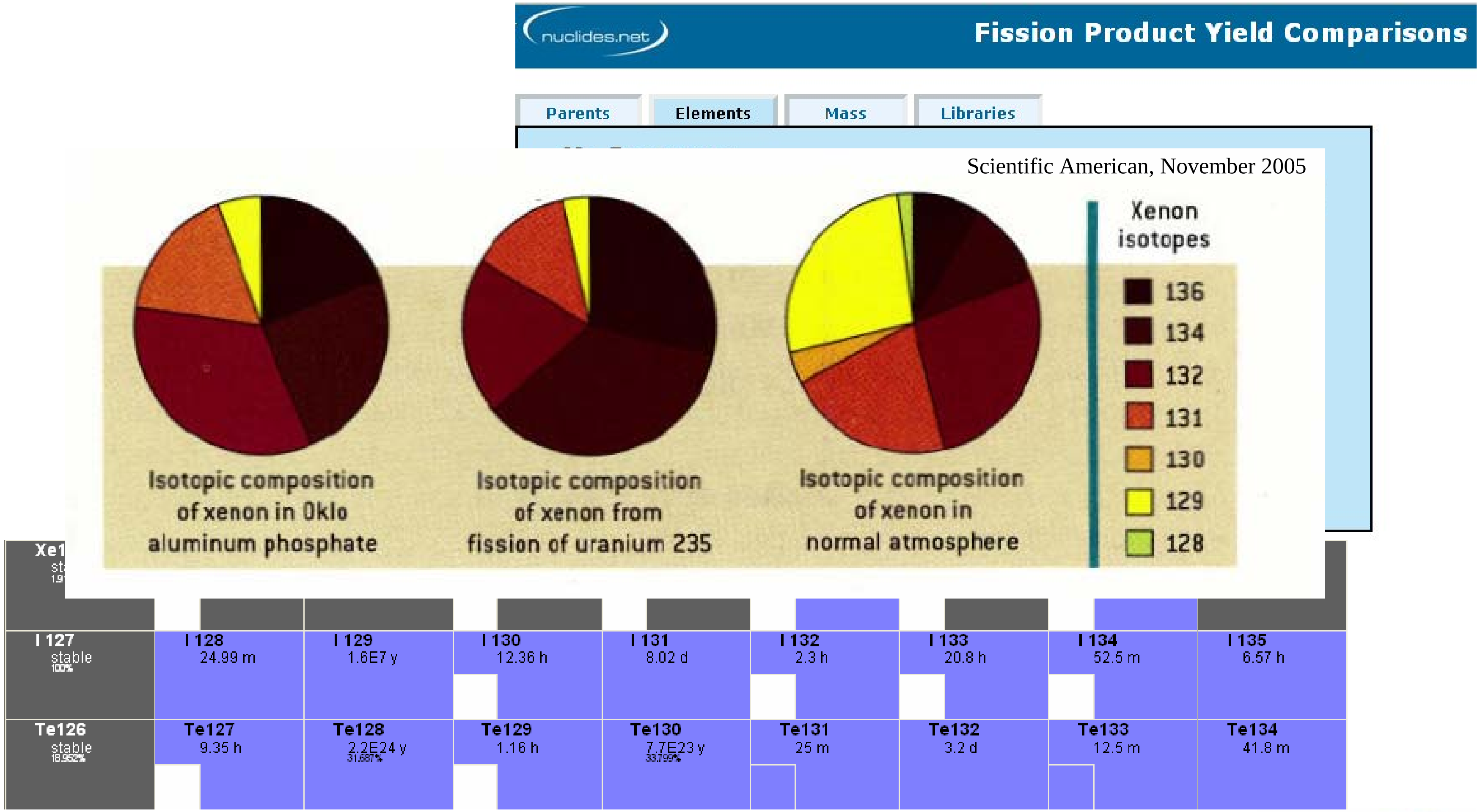
Analysis of Xenon Composition (II)



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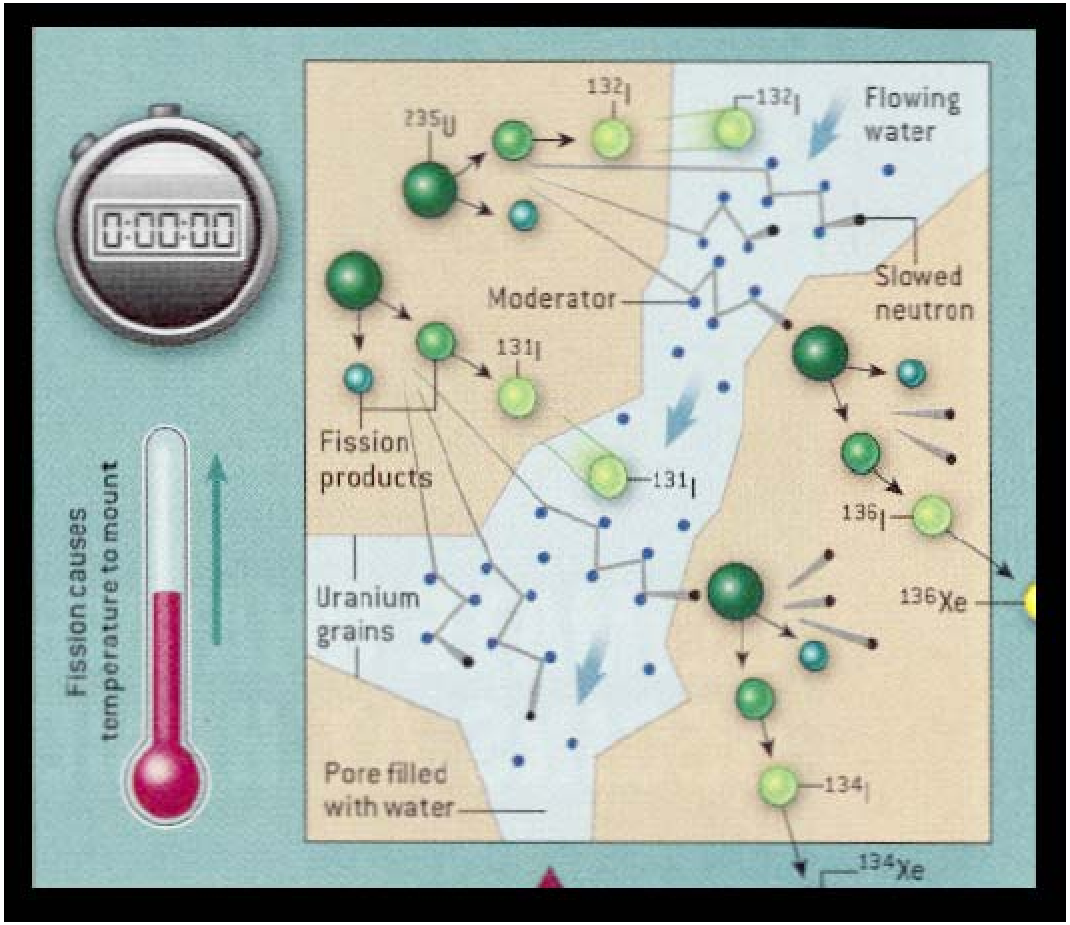
A Nuclear Geyser (I)

- Reactor operation was cyclical.
- Groundwater flowing through reactor zone moderates neutrons and starts chain reaction on ^{235}U.
- Xenon and Iodine fission products are mostly washed away in the groundwater.



Scientific American, November 2005

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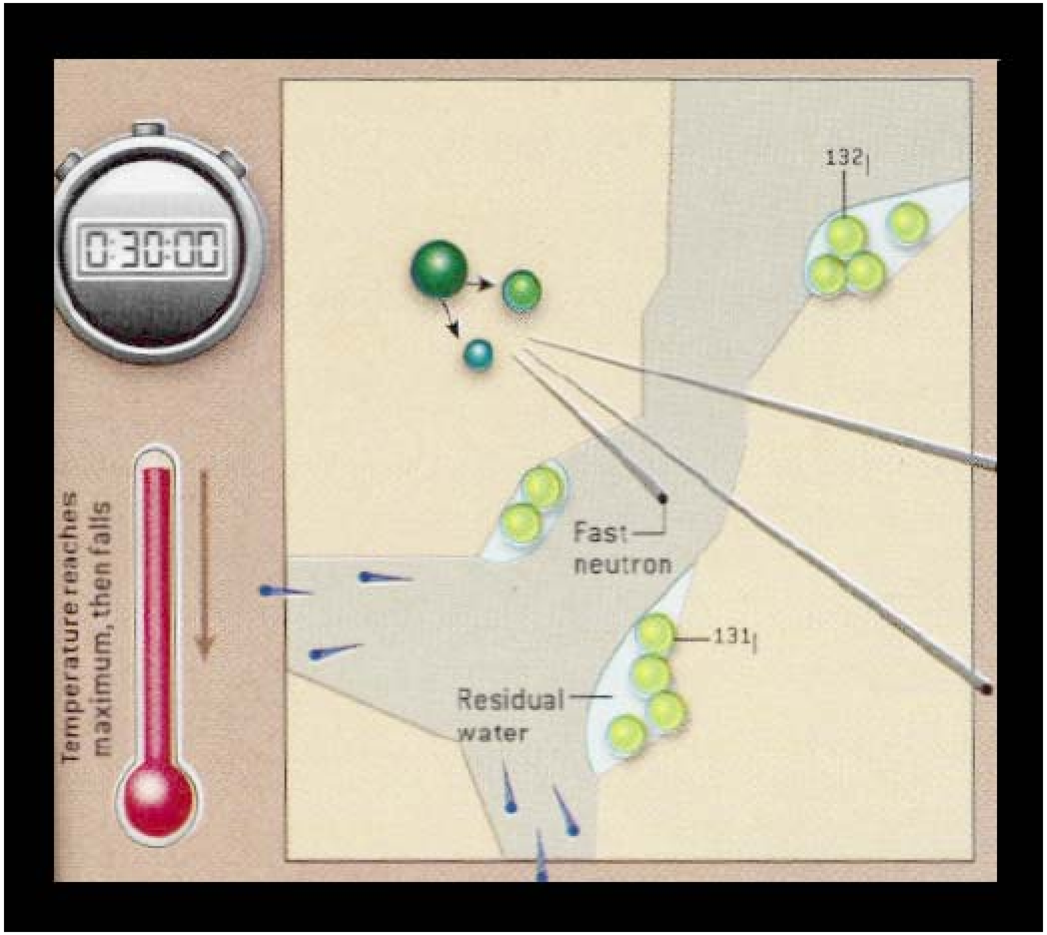
A Nuclear Geyser (II)

- After a period of 30 minutes, most of the groundwater has been boiled off.
- No moderation means that reactor operation shuts down.
- Some Iodine fission products are retained as the temperature starts to fall.



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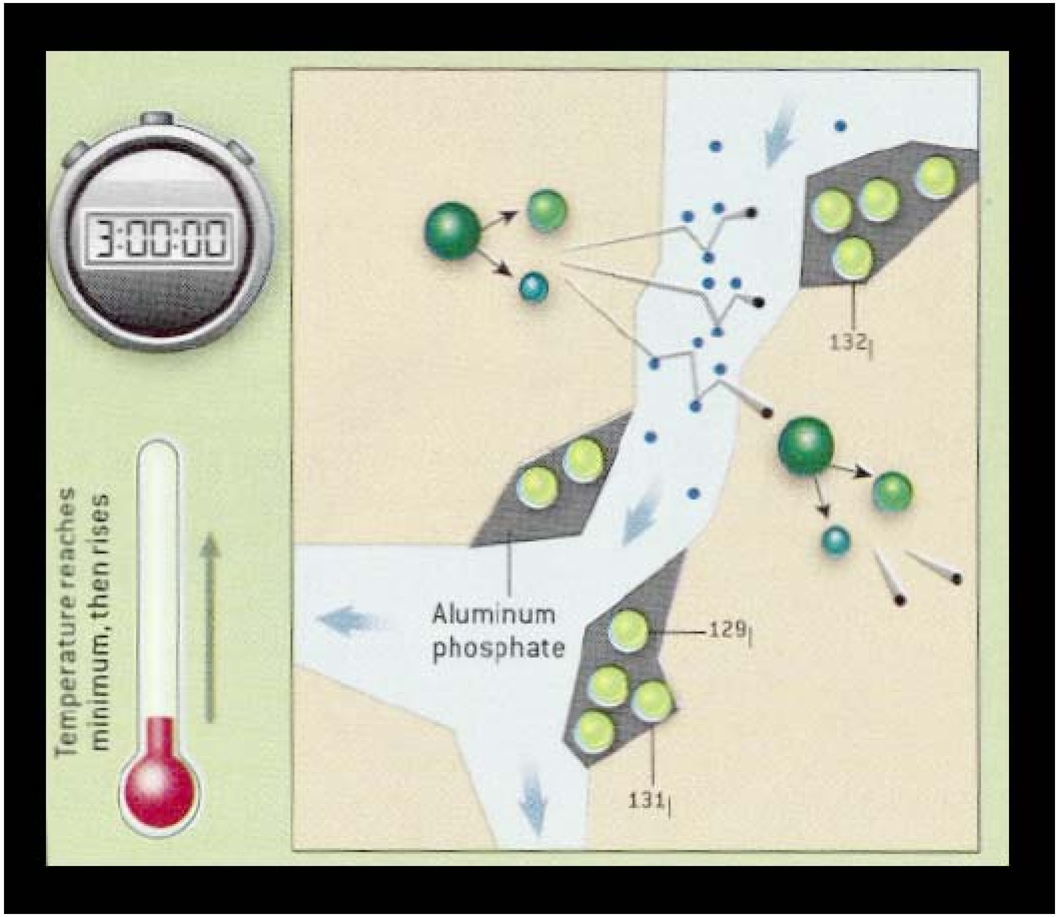
A Nuclear Geyser (III)

- Groundwater returns to reactor zone after a few hours as temperature returns to normal.
- Aluminium Phosphate minerals are formed which trap Iodine fission products.
- These then decay in the minerals to stable Xenon isotopes and remain there for 2 billion years.
- At the same time, reactor operation begins again.
- The cycle is believed to have continued for 150,000 years



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Fission Yields and the Oklo Reactor

Summary/Perspectives

- Revolutionary discovery at Oklo mines in Gabon that nuclear fission is, in fact, a completely natural process.
- A nuclear forensic analysis of the fission products contained in samples allow reconstruction of a reactor which operated 2 billion years ago with quite remarkable precision.
- The reactor is believed to have operated in 30 minute pulses with 2.5 hour dormant periods for over 150,000 years.
- Further analysis of Oklo samples allow:
  - Studies concerning the storage of nuclear wastes in natural environments over extremely long time periods.
  - Verification of the variability of the physical constants of nature.
- This be a common phenomenon: speculation about sites in Colorado, Canada and Australia.

Fission Yields and the Oklo Reactor

References

- Meshik, Hohenberg & Pravdivtseva, "Record of Cycling Operation of the Natrual Nuclear Reactor in the Oklo/Okelobondo Area in Gabon," Phys. Rev. Lett. 93, 182302-1 (2004).
- Meshik, A., "The Workings of an Ancient Nuclear Reactor," Scientific American, November 2005.
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- Karam, A, "The Natural Nuclear Reactor at Oklo: A comparison with Modern Nuclear Reactors,"(2005) http://www.physics.isu.edu/radinf/Files/Okloreactor.pdf
- Holloway, R., "Plutonium fission in the Oklo natural reactor," Savannah River DS-MS-80-75, (http://sti.srs.gov/fulltext/dpms8075/dpms8075.pdf).

Question 1: Iodine production through fission

A sample of ^{235}U has been irradiated in a thermal reactor. A total of 10^7 fissions have been measured. The irradiated sample is stored for 1 day. The safety officer detects traces of iodine ^{131}I (a volatile element) in his filters.

What is the activity and dose rate of the iodine present in the sample one day after irradiation? Use the FY yield module and the decay module.

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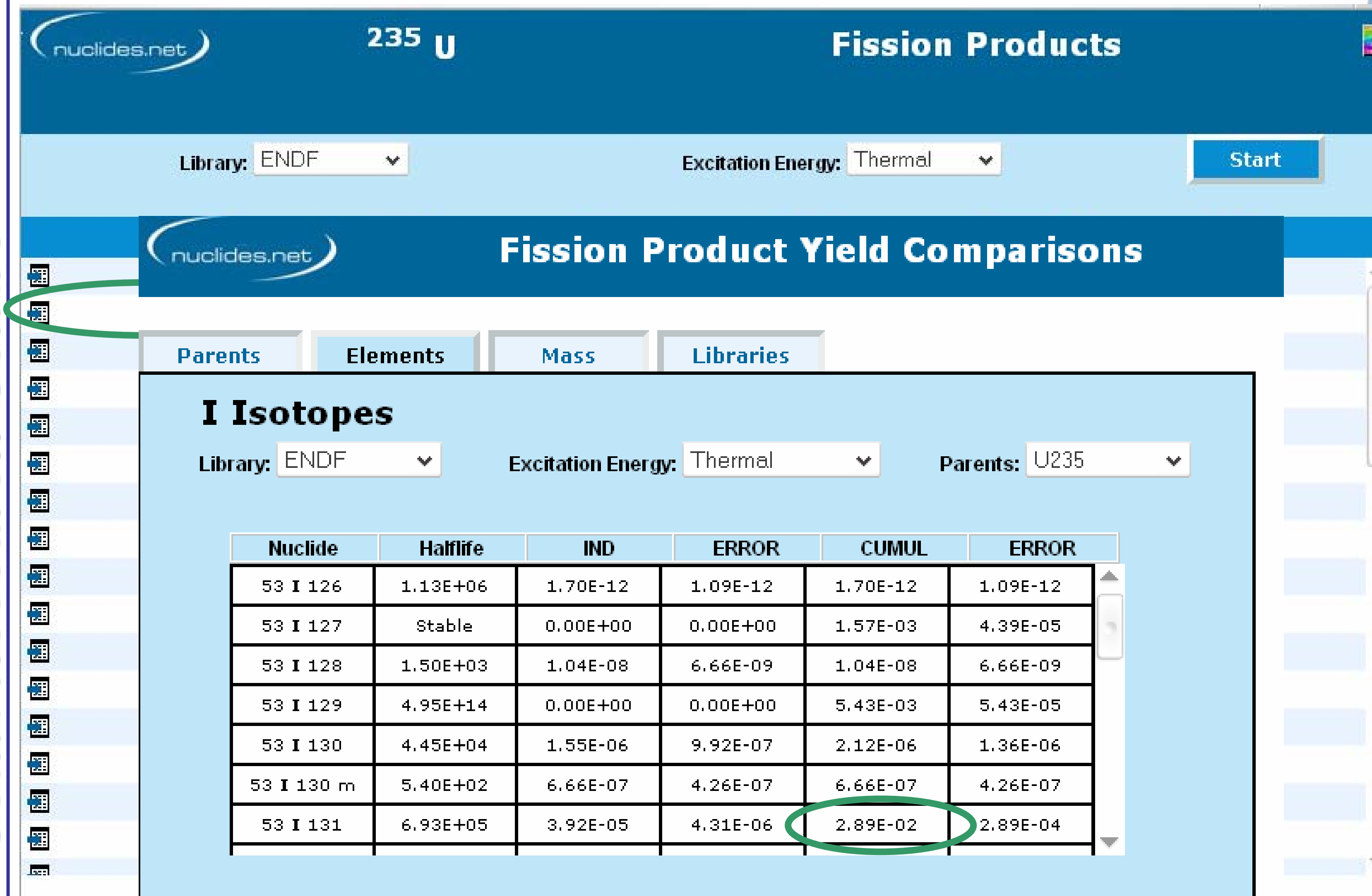

Nuclide	Halflife	IND	ERROR	CUMUL	ERROR
53 I 126	1.13E+06	1.70E-12	1.09E-12	1.70E-12	1.09E-12
53 I 127	Stable	0.00E+00	0.00E+00	1.57E-03	4.39E-05
53 I 128	1.50E+03	1.04E-08	6.66E-09	1.04E-08	6.66E-09
53 I 129	4.95E+14	0.00E+00	0.00E+00	5.43E-03	5.43E-05
53 I 130	4.45E+04	1.55E-06	9.92E-07	2.12E-06	1.36E-06
53 I 130 m	5.40E+02	6.66E-07	4.26E-07	6.66E-07	4.26E-07
53 I 131	6.93E+05	3.92E-05	4.31E-06	2.89E-02	2.89E-04

Iodine after irradiation

$$N_{I^{131}} = 10^7 \cdot 0.0289 = 2.89 \cdot 10^5 \; atoms$$

nuclides.net 131 I **Full Decay**

Options

Quantity: Number(atoms) 2.89E5 Distance(cm): 100 Min. Prod.: 1E-02 Start

Time: Days 1 Number of time steps: 1 N° Chains: 1 Reset

Parent+Daughters	N(atoms)	M(g)	A(Bq)	G(keV/s)	dH/dt(µSv/hr)
53 I131	2.6507E+05	5.7620E-17	2.6513E-01	1.0076E+02	1.9291E-08
54 Xe131 m	2.3230E+04	5.0495E-18	1.5740E-02	5.0573E-02	1.9948E-09
54 Xe131 Stable	6.9669E+02	1.5144E-19	0.0000E+00	0.0000E+00	0.0000E+00
Total :	2.8900E+05	6.2821E-17	2.8087E-01	1.0081E+02	2.1286E-08

Question 2: Thermal Fission in Oklo

Suppose there was 500 kg of uranium ore in a particular reactor zone of the Oklo natural nuclear reactor. If the average thermal neutron flux through this zone was 10^6 cm^{-2}s^{-1} (remembering that the ^{235}U enrichment 2 billion years ago was 3%):

- How many thermal fission reactions took place in one second?
- How many thermal fissions in a reactor pulse (30 min duration)?
- Neglecting production through decay, how much ^{131}I and ^{132}I would have been produced in this half-hour period? (Use ENDF library.)
- What is the quantity of these isotopes that will remain when the next reactor pulse starts (2.5 hours later)?

Hint: Remember that 1 barn = 10^{-24} cm^2 and that the number of reactions per second = $N\Phi\sigma$. Use the cross section data, fission yield module and decay calculator in Nuclides.net..

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Question 2: Thermal Fission in Oklo

(a) Mass of ^{235}U = 500 kg * 0.03 = 15 kg

Number of ^{235}U Nuclei (use mass-activity converter) N = ***3.8.10^{25}***

Cross section (thermal, 2200 m/s) σ = ***583 b*** = 5.83.10^{-22} cm^2

Neutron Flux Φ = 1.10^6 cm^{-2}s^{-1}

Number of reactions per second = $N\Phi\sigma$ = 2.2.10^{10}

(b) Number of reactions in reactor pulse
= 30 * 60 * 2.2.10^{10} = 3.96.10^{13}

(c) Independent fission yields (ENDF)

^{131}I yield = ***3.92.10^{-5}*** * 3.96.10^{13} = 1.55.10^9 atoms

^{132}I yield = ***9.13.10^{-5}*** * 3.96.10^{13} = 3.62.10^9 atoms

(d) Decay after 2.5 hours:

^{131}I after decay = ***1.54.10^9*** atoms

^{132}I after decay = ***1.70.10^9*** atoms